

# Integer Quantum Hall Effect of Interacting Electrons in Graphene

Xin-Zhong Yan<sup>1</sup> and C. S. Ting<sup>2</sup>

<sup>1</sup>*Institute of Physics, Chinese Academy of Sciences, P.O. Box 603, Beijing 100190, China*

<sup>2</sup>*Texas Center for Superconductivity, University of Houston, Houston, Texas 77204, USA*

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By taking into account the charge and spin orderings and the exchange interactions between all the Landau levels, we investigate the integer quantum Hall effect of electrons in graphene using the mean-field theory. At the fillings  $\nu = 4n + 2$  with  $n = 0, 1, \dots$ , the system is in the high-symmetry state with the Landau levels four-fold degenerated. We show that with doping the degenerated lowest empty levels can be sequentially filled one level by one level; the filled level is lower than the empty ones because of the symmetry breaking. This result explains the step  $\Delta\nu = 1$  in the integer quantized Hall conductivity of the experimental observations. We also present in the supplemental material a high efficient method for dealing with huge number of the Coulomb couplings between all the levels.

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The study of quantum Hall effect (QHE) is an important aspect of the graphene physics. By the noninteracting electron model, the Hall conductivity is given by  $\sigma_{xy} = \nu e^2/h$  and  $\nu = \pm(4n + 2)$  with  $n = 0, 1, \dots$  as the index of highest-occupied Landau level (LL) as observed in the very early experiments [1–3]. The step  $\Delta\nu = 4$  stems from the spin and valley degeneracy of LLs of electrons in graphene. In addition to the fillings  $\nu = \pm 2, \pm 6, \dots$ , the experiments then observed the states of 0,  $\pm 1$ , and  $\pm 4$  at strong magnetic field [4, 5]. The appearance of these states were attributed to various reasons including the long-range interactions between electrons [6, 7] with ferromagnetization [8] or with disorders [9], antiferromagnetic spin ordering and charge-density wave due to short-range interactions [10], the existence of excitonic gap [11], and the field dependent Peierls distortion [12]. It is considered that the four-fold degeneracy of the LLs is lifted due to these factors. Later, all the integer factors of  $|\nu| \leq 10$  in high-quality suspended graphene even at weak field ( $< 1$  T) were observed [13]. For the clean system at weak field, it seems that only the Coulomb interactions between electrons can be considered to explain the experiment. However, it can be shown that the four-fold degeneracy cannot be completely lifted at  $\nu = \pm(4n + 2)$  because of system's symmetry. Therefore, the experimental observation is a mystery.

In this Letter, using the mean-field theory (MFT), we show the filling factor varies as  $\Delta\nu = 1$  for the integer QHE (IQHE) of electrons in graphene. For the interacting electrons, the LLs are not rigid but vary with the electron doping; the highest-occupied level is always lower than the lowest-unoccupied level though they might be originally degenerated before the doping. On the other hand, we intend to develop a high-efficient method for dealing with the Coulomb couplings between all the LLs. At weak magnetic field, the treatment of inter-electronic interactions within the single level as in the conventional theory is insufficient. Since there are huge-number couplings at magnetic field  $< 1$  T, the numerical computa-

tion is formidable. To overcome the numerical difficulty, we present the high-efficient method in the supplemental material (SM).

We begin with the description of the electron system in graphene. The honeycomb lattice of graphene shown in Fig. 1 (left) contains atoms  $a$  and  $b$  with lattice constant  $a_0 \approx 2.4$  Å. The Hamiltonian of the electrons with a neutralizing background is

$$H = -t \sum_{\langle ij \rangle s} c_{is}^\dagger c_{js} + U \sum_j \delta n_{j\uparrow} \delta n_{j\downarrow} + \frac{1}{2} \sum_{i \neq j} v_{ij} \delta n_i \delta n_j$$

where  $c_{is}^\dagger$  ( $c_{is}$ ) creates (annihilates) an electron of spin  $s$  in site  $i$ ,  $\langle ij \rangle$  means the sum over the nearest-neighbor (NN) sites,  $t \approx 3$  eV is the NN hopping energy [14, 15],  $\delta n_{is} = n_{is} - n_s$  is the number deviation of electrons of spin  $s$  at site  $i$  from the average occupation  $n_s$ , and  $U$  and  $v_{ij}$  are the Coulomb interactions between electrons. In real space,  $v_{ij} = v(r_{ij})$  with  $r_{ij}$  the distance between sites  $i$  and  $j$  is given by  $v(r) = e^2[1 - \exp(-q_0 r)]/r$  where  $q_0$  is a parameter taking into account the effect of wave function spreading. We here take  $q_0 = 0.5/a_0$ .

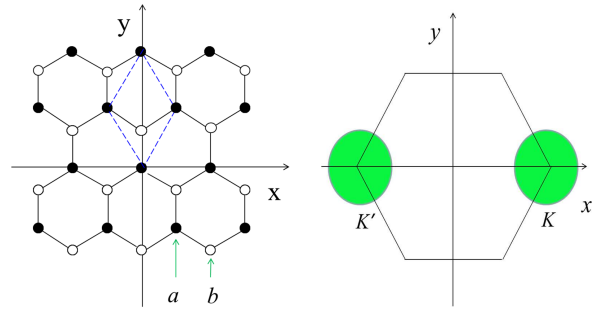


FIG. 1. (color online) Left: Lattice structure of graphene contains atoms  $a$  (black) and  $b$  (white). The dashed diamond is the unit cell. Right: First Brillouin zone and the two valleys  $K$  and  $K'$  in the momentum space.

We here treat the interactions between electrons by the MFT. In the Hartree term, there are spin and charge orderings with the order parameters defined as  $m_j = (\langle \delta n_{j\uparrow} \rangle - \langle \delta n_{j\downarrow} \rangle)/2$  and  $\rho_j = (\langle \delta n_{j\uparrow} \rangle + \langle \delta n_{j\downarrow} \rangle)$ , respectively. These parameters depend only on the index  $l$  ( $= a$  or  $b$ ) of the sublattice,  $m_j = m_l$  and  $\rho_j = \rho_l$ , where the position  $j$  belongs to the sublattice  $l$ . There is only one parameter  $\rho_a = -\rho_b \equiv \rho$  for the charge ordering because of the charge neutrality. For the Fock term, we take the screening effect (due to the charge-density fluctuations) in the exchange interaction by the Thomas-Fermi (TF) screening function  $\epsilon(q) = 1 + q_{TF}/q$  with  $q_{TF}$  as the TF wave number and  $q$  the momentum transfer between electrons. By translating the lattice to the continuous space, the Hamiltonian under the MFT is obtained as (see SM)

$$H = \sum_{vs} \left[ \int d\vec{r} C_{vs}^\dagger(r) h_v(\vec{p}) C_{vs}(r) + \int d\vec{r} \int d\vec{r}' C_{vs}^\dagger(r) \Sigma^{vs}(r, r') C_{vs}(r') \right] \quad (1)$$

where  $h_v(\vec{p}) = v_0(s_v p_x \sigma_1 + p_y \sigma_2)$  with  $\vec{p}$  the momentum operator and  $v_0 = \sqrt{3}ta_0/2\hbar$  as the Fermi velocity,  $s_v = 1$  ( $-1$ ) for electrons in valley  $v = K$  ( $K' = -K$ ) [see Fig.1 (right)], the Pauli matrices  $\sigma$ 's operate in the space ( $a, b$ ) of sublattices,  $C_{vs}^\dagger(r) = [c_{avs}^\dagger(r), c_{bvs}^\dagger(r)]$  with  $c_{a(b)vs}^\dagger(r)$  creating an electron of spin  $s$  of valley  $v$  on position  $r$  in  $a$  ( $b$ ) sublattice, and  $\Sigma^{vs}(r, r')$  is a  $2 \times 2$  matrix of the self-energy. We will use the units in which  $\hbar = a_0 = v_0 = 1$ . The energy unit then is  $\epsilon_0 = \hbar v_0/a_0 = 1$ .

Under a magnetic field  $B$  perpendicular to the graphene plane, we take the vector potential as  $\vec{A}(r) = (0, Bx)$ . The momentum along  $y$ -direction is a good number and is denoted as  $k$ . For going to the LL picture, we expand the operator  $C_{vs}(r)$  as

$$C_{vs}(r) = \sum_{nk\lambda} \phi_{nk}^v(r) \psi_\lambda^{vs}(n) \hat{a}_{\lambda nk}^{vs} \quad (2)$$

where  $\phi_{nk}^v(r)$  is a  $2 \times 2$  matrix given by

$$\phi_{nk}^{v=K}(r) = \frac{e^{iky}}{\sqrt{L}} \begin{pmatrix} \phi_{n-1}(x-x_c) & 0 \\ 0 & i\phi_n(x-x_c) \end{pmatrix},$$

and  $\phi_{nk}^{K'}(r) = i\sigma_1 \phi_{n,-k}^{K*}(r) \sigma_1$ , with  $L$  the length of the lattice along  $y$  direction and  $\phi_n(x-x_c)$  the  $n$ th level wave function of a harmonic oscillator along  $x$  direction with the center at  $x_c = -k/B$ ,  $\psi_\lambda^{vs}(n)$  (real) is a two-component spinner, and  $\hat{a}_{\lambda nk}^{vs}$  annihilates an electron of momentum  $k$  (along  $y$  direction) and spin  $s$  in valley  $v$  at  $\lambda$ th LL of index  $n$ . For  $n < 0$ ,  $\phi_n$  is understood as 0. In the LL picture, the elements of the self-energy matrix is given by (see SM)

$$\Sigma_{ll'}^{vs}(n) = (v_c \rho_l - sU m_l) \delta_{ll'} - \sum_{n'} v_{ll'}^v(n, n') g_{ll'}^{vs}(n'), \quad (3)$$

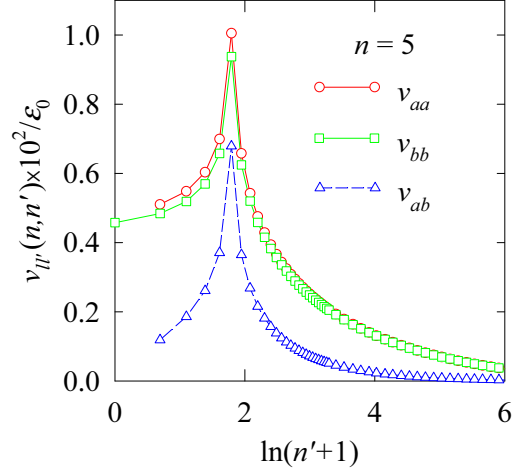


FIG. 2. (color online)  $K$ -valley interaction  $v_{ll'}(n, n')$ 's as functions of  $n'$  at  $n = 5$  and  $B = 0.5$  T and  $\nu = 0$ .  $v_{aa}(n, n')$  and  $v_{ab}(n, n')$  are defined for  $n' \geq 1$ .

with  $v_c$  a potential for charge ordering and  $s = 1$  ( $-1$ ) for spin up (down), and the two matrices  $v^v(n, n')$  and  $g^{vs}(n')$  are given by

$$\begin{aligned} v^K(n, n') &= \sigma_1 v^{K'}(n, n') \sigma_1 \\ &= \int_0^\infty \frac{q dq}{2\pi} v^{sc}(q) e^{-\xi} \xi^m J_{n, n'}(\xi) \otimes J_{n, n'}^t(\xi) \\ J_{n, n'}^t(\xi) &= \left[ \sqrt{\frac{(n_2 - 1)!}{(n_1 - 1)!}} L_{n_2 - 1}^m(\xi), \sqrt{\frac{n_2!}{n_1!}} L_{n_2}^m(\xi) \right] \\ g^{vs}(n) &= \sum_\lambda [f_\lambda^{vs}(n) - 1/2] \psi_\lambda^{vs}(n) \otimes \psi_\lambda^{vs, t}(n) \end{aligned}$$

where  $v^{sc}(q) = v(q)/\epsilon(q)$  is the screened interaction,  $\xi = q^2/2B$ ,  $J_{n, n'}^t$  ( $\psi_\lambda^{vs, t}$ ) is a transpose of  $J_{n, n'}$  ( $\psi_\lambda^{vs}$ ),  $L_{n_2}^m(\xi)$  is the Laguerre polynomial,  $n_1 = \max(n, n')$ ,  $n_2 = \min(n, n')$ ,  $m = |n - n'|$ , and  $f_\lambda^{vs}(n)$  is the Fermi distribution of valley- $v$  and spin- $s$  electrons in the  $\lambda$ th level of index  $n$ . The LL  $E_{\lambda n}^{vs}$  is determined by

$$[\sqrt{2Bn} \sigma_1 + \Sigma^{vs}(n)] \psi_\lambda^{vs}(n) = E_{\lambda n}^{vs} \psi_\lambda^{vs}(n). \quad (4)$$

The charge and spin orders are calculated by

$$\rho = \frac{s_0 B}{4\pi} \sum_{l\lambda nvs} s_l f_\lambda^{vs}(n) |\psi_{l\lambda}^{vs}(n)|^2 \quad (5)$$

$$m_l = \frac{s_0 B}{4\pi} \sum_{\lambda nvs} s f_\lambda^{vs}(n) |\psi_{l\lambda}^{vs}(n)|^2 \quad (6)$$

where  $s_0 = \sqrt{3}/2$  is the area of the unit cell,  $B/2\pi$  is the spacial degeneracy of the Landau state,  $\psi_{l\lambda}^{vs}(n)$  is the  $l$ th component of  $\psi_\lambda^{vs}(n)$  and  $s_l = 1$  ( $-1$ ) for  $l = a$  ( $b$ ). By taking the Hubbard  $U/\epsilon_0 = 2.5$  [16], the potential  $v_c$  is then determined as  $v_c/\epsilon_0 = 0.173$ .

Here, the magnetic field is in unit of  $B_0 = \hbar c/ea_0^2 = 1.105 \times 10^4$  T. Corresponding to the momentum cutoff

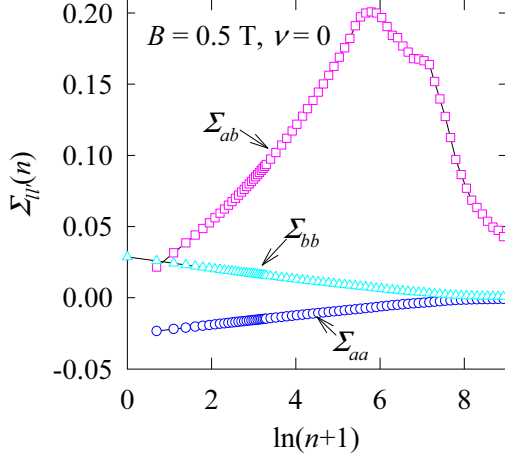


FIG. 3. (color online) Self-energy  $\Sigma_{ll}(n)$ 's of spin-up electrons in valley  $K$  as functions of  $n$  at  $B = 0.5$  T and  $\nu = 0$ .  $\Sigma_{aa}(n)$  and  $\Sigma_{ab}(n)$  are defined for  $n \geq 1$ .

$p_c \sim 1$ , the largest Landau index is  $N \sim 0.5/B$ . For  $B = 0.5$  T  $= 0.45 \times 10^{-4} B_0$ , we have  $N \sim 1.1 \times 10^4$ . According to Eq. (3), the number of the  $2 \times 2$  interaction matrix  $v^v(n, n') = v^v(n', n)$  in the calculation is about  $N^2/2$ . Because each element of the matrix is obtained by the integral with the Laguerre polynomials involved, for large  $n_2 = \min(n, n')$ , the calculation for the element is very time-consuming. It is therefore a formidable work to numerically calculate  $N^2/2$  matrices at weak  $B$ . Most of the works simplify the calculation by taking only one term of  $n' = n$  in the sum in Eq. (3). In Fig. 2,  $v_{ll}(n, n') \equiv v_{ll}^K(n, n')$ 's are shown as functions of  $n'$  at  $n = 5$  and  $B = 0.5$  T and  $\nu = 0$ . As seen from Fig. 2, the interactions  $v_{ll}^K(n, n')$ 's vary slowly with  $n'$  at a given  $n$ . Therefore, the only peak-value term in a calculation is not sufficient for reflecting the interaction effect. To resolve the problem, we have developed a high efficient method for the series sum (see SM).

We have self-consistently solved the Eqs. (3)-(6). Shown in Fig. 3 are the self-energy  $\Sigma_{ll}(n) \equiv \Sigma_{ll}^{K\uparrow}(n)$  at  $B = 0.5$  T and  $\nu = 0$ . In valley  $K$ , the self-energy elements  $\Sigma_{aa}^{K\uparrow}(n)$  and  $\Sigma_{ab}^{K\uparrow}(n)$  are defined for  $n \geq 1$ . The element  $\Sigma_{ab}^{K\uparrow}(n)$  describes the exchange effect due to interactions between the sublattice  $a$  and  $b$ . Because of the induction by the inter-sublattice hopping,  $\Sigma_{ab}^{K\uparrow}(n)$  is stronger than  $\Sigma_{aa}^{K\uparrow}(n)$  and  $\Sigma_{bb}^{K\uparrow}(n)$  for  $n > 1$ . On the other hand,  $\Sigma_{aa}^{K\uparrow}(n)$  and  $\Sigma_{bb}^{K\uparrow}(n)$  vary slowly with the index  $n$ . At the charge neutrality point (CNP)  $\nu = 0$ ,  $\Sigma_{ll}^{K\uparrow}(n)$  at small  $n$  comes mainly from the spin ordering term  $-sUm_l$ .

Our main result is shown in Fig. 4 where the LLs at filling numbers  $\nu = 0$  to 6 are depicted for  $B = 0.5$  T. At  $\nu = 0$ , the four-fold degeneracy of the levels of  $n = 0$  is partially lifted due to the antiferromagnetic spin order-

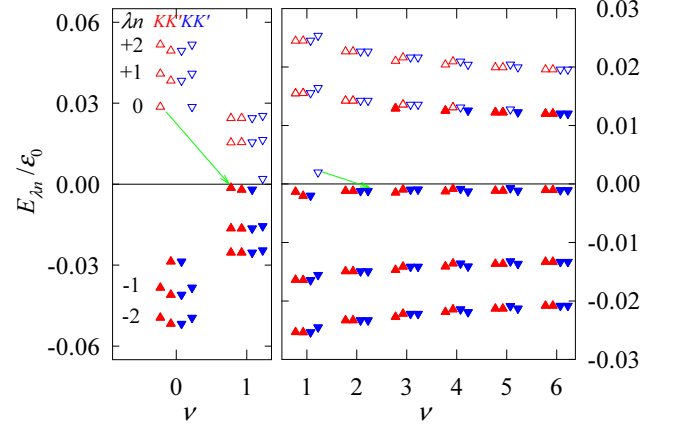


FIG. 4. (color online) Landau levels  $E_{\lambda n}$  at  $B = 0.5$  T. The red-filled (empty) upward-triangles for occupied (empty) levels of spin-up electrons in valley  $K$  and  $K'$ . The blue downward-triangles for the corresponding levels of spin-down electrons. In each column corresponding to a spin and valley at filling number  $\nu$ , the five levels are for  $n = 0$  and  $\pm n$  ( $n = 1, 2$ ) with  $+n$  ( $-n$ ) standing for upper (lower) energy level of index  $n$ . An arrow indicates the energy change of a level from empty to filled state.

ing (see Fig. 5) with a gap between the empty and filled levels. The two highest filled levels are the levels of  $n = 0$  for spin-up electrons in valley  $K'$  (occupying sublattice  $a$ ) and spin-down electrons in valley  $K$  (occupying sublattice  $b$ ), respectively. However, because of  $\rho = 0$  and  $\sigma_1 \Sigma^{Ks}(n) \sigma_1 = \Sigma^{K'\bar{s}}(n)$  with  $\bar{s} = -s$ , each level still has a degeneracy 2. At the CNP, the levels satisfy the particle-hole symmetry  $[\sigma_2 \Sigma^{Ks}(n) \sigma_2 = -\Sigma^{K's}(n)]$ : for a positive  $K$ -valley level, there is a negative  $K'$ -valley level of the same spin electrons.

The question now is, with doping electrons, should the two empty levels of  $n = 0$  be filled or can there exist a state only one of them filled? The former will give a quantum Hall state of  $\nu = 2$  and the latter be  $\nu = 1$ . Our result shows that there can exist a state with the  $K$  (or  $K'$ ) valley level of  $n = 0$  filled with spin-up (down) electrons [occupying sublattice  $b$  ( $a$ )]. The energy of this occupied level (for the case  $\nu = 1$  shown in Fig. 4) is  $\Sigma_{bb}^{K\uparrow}(0) = v_c \rho_b - Um_b + \Sigma_{bb}^{K\uparrow, xc}(0)$  where the last term is the exchange part of the self-energy. The empty level is  $\Sigma_{aa}^{K'\downarrow}(0) = v_c \rho_a + Um_a + \Sigma_{bb}^{K'\downarrow, xc}(0)$ . At this state, above the spin ordering there is another symmetry breaking from the charge ordering. Since the sublattice  $b$  now is occupied with more electrons than the sublattice  $a$ , the charge order parameter is obtained as  $\rho = \rho_a = -\rho_b < 0$  as shown in Fig. 5. Meanwhile, the spin ordering in sublattice  $b$  is weakened with magnitude  $|m_b|$  smaller than  $|m_a|$ . The charge ordering gives rise to a negative contribution to the gap  $\Sigma_{aa}^{K'\downarrow}(0) - \Sigma_{bb}^{K\uparrow}(0)$  which means it is not favored by the Coulomb interactions. However, since

the spin ordering is still strong at this state, the energy cost of the charge ordering is fully compensated by the spin ordering. In addition, the difference in the exchange part gives rise to significant positive contribution to the gap. As a result, the imbalanced electron distributions in the two valleys and in two spins lead to the filled level lower than the empty one. With further doping, the only empty level in valley  $K'$  for spin-down of  $n = 0$  will be filled with electrons, giving rise to the state of  $\nu = 2$ .

In the state of  $\nu = 2$ , the orderings vanish  $\rho_l = m_l = 0$  and the self-energy satisfies the symmetry  $\sigma_1 \Sigma^{vs}(n) \sigma_1 = \Sigma^{\bar{v}s}(n)$  with  $\bar{v} = -v$  and becomes independent of  $s$ . Therefore, the state of  $\nu = 2$  is a high symmetry state with degeneracy 4. We again go back to the similar question as mentioned above, with further doping electrons, should the four empty levels of  $n = 1$  all be filled or can there exist a state only one of them filled? The answer is the four levels can be sequentially filled one level by one level. Though they are degenerated at  $\nu = 2$ , the filled levels are lower than the empty levels which can be seen from Fig. 4. For  $\nu > 2$ , since the orderings are weak, the changes in the levels stem predominately from the exchange interactions. For  $\nu = 3$  and 5, since more levels are occupied with the spin-up electrons, we get the ferromagnetic states with unequal magnitudes of spin orderings in the two sublattice as shown in Fig. 5. Corresponding to the imbalanced occupation in the two valleys and thereby in the two sublattices, the charge ordering is finite at  $\nu = 3$  and 5. At  $\nu = 4$  where the levels for spin-up electron in valley  $K$  and spin-down electron in valley  $K'$  are filled, the symmetry  $\sigma_1 \Sigma^{vs}(n) \sigma_1 = \Sigma^{\bar{v}s}(n)$  leads to a degeneracy 2. Because the numbers of electrons occupied in the two sublattice are equal, the charge ordering vanishes. But the spins in two sublattice are not balanced, the system is in the antiferromagnetic state. At  $\nu = 6$ , all the upper four levels of  $n = 1$  are filled and we reach again a high symmetric state with  $\rho = m_l = 0$  as in  $\nu = 2$ .

At higher filling numbers  $\nu > 6$ , the level filling processes are similar as that from  $\nu = 2$  to 6. But the gap between the highest filled level and the lowest empty level of an index  $n$  decreases with  $n$  because the Coulomb coupling is less important at high  $n$ . Therefore, above certain large  $\nu$ , the step  $\Delta\nu = 4$  in the Hall conductivity will be observed.

We note that all the levels vary with doping as clearly seen from Fig. 4. When a level is filled with doping, the level not only shifts its self, but also influences other levels through the Coulomb interactions.

It should be indicated that a constant term reflecting the gate voltage should be included in the square brackets in Eq. (4) for determining the absolute values of the levels. This term is given by  $e^2 n_0 \nu / C \epsilon_0$  where  $-e$  is the charge of an electron,  $n_0 = \sqrt{3}B/8\pi$  is the number of doped electrons per atom per level, and  $C$  is the charge capacity per atom. The magnitude of  $C$  is dependent on

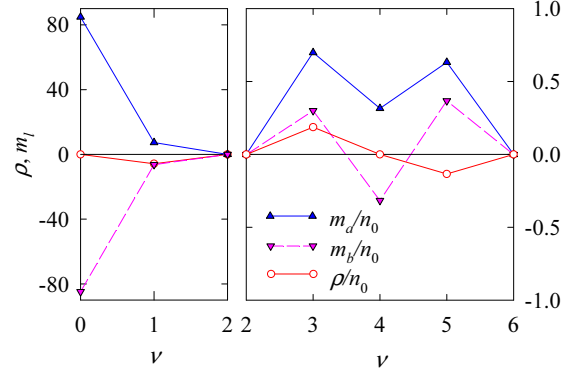


FIG. 5. (color online) Spin and charge ordering parameters as functions of filling number  $\nu$ . Here,  $\rho \equiv \rho_a = -\rho_b$ , and  $n_0 = \sqrt{3}B/8\pi$  is the doped electrons per level per atom.

the real system and is smaller than (or the same order)  $2 \times 10^{-5} e/V$  of the system the graphene placed on a  $\text{SiO}_2$  substrate [1]. This term increases with  $\nu$  much stronger than the decreasing of the levels shown in Fig. 4. As a result, the absolute values of the levels increase with the electron doping.

We have neglected the Zeeman splitting. At  $B = 0.5$  T, the splitting  $\mu_B B / \epsilon_0 = 1.1 \times 10^{-5}$  is much less than the smallest gap  $\sim 5 \times 10^{-4}$  ( $= 1.3$  meV) appeared at  $\nu = 5$  shown in Fig. 4.

Since the system satisfies the particle-hole symmetry [17], the above results for the electron doping can be converted to the case of hole doping.

In conclusion, we have investigated IQHS of electrons in graphene by the MFT taking into account the Coulomb couplings between all the Landau levels. At the fillings  $\nu = 4n + 2$ , there are no spin and charge orderings and the Landau levels are four-fold degenerated because of the invariance of the system under the exchanges of spin and valley. We have shown that the lowest degenerated empty levels can be sequentially filled one level by one level with the filled levels lower than the empty levels. The thing can happen because with doping the inter-electronic interactions give rise to the spin and valley dependent self-energy and the symmetry is broken. This filling process results in a step of  $\Delta\nu = 1$  in the quantized Hall conductivity.

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